

Fragments formation in proton-induced fission at 660 MeV of ^{238}U

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In this paper the experimental data regarding ^{238}U fission at proton energy 660 MeV are presented and analyzed. The absolute values of 51 cross-sections of fragment production, in the range of mass numbers 7-70 amu, are obtained and evinced. Such fragments may be considered as a third particle accompanying two other fragments. The ternary fragment mass distribution may bring informations on the nucleon composition of the neck, and the high energy associated to the fissioning nucleus might provide an overlap between the ternary and binary fission, in the mass region of fission fragments 50-70 amu. Furthermore, the cross-section of ^{238}U ternary fission is determined, as well as the ratio for binary/ternary fission, in order to obtain new prominent data delving into the underlying fission mechanism concerning the proton-induced fission at intermediate energies.

1 Introduction

The investigation on nuclear fission reactions, induced by nucleons on heavy targets, can be associated with two fundamental questions, which are not fully explored: the reaction mechanism induced by neutrons and protons for a wide range of energy, and nuclear fission properties. They are somehow related, since the fission mechanism is determined by static and dynamic features of nuclear matter during the global restructuring of the compound nucleus, before the separation into two or more fragments. The phenomena encompassing nuclear fission have been intensely investigated, in the last 70 years, from the experimental as well as the theoretical point of view. The wide interest related to those phenomena is associated with their relevant practical applications and important scientific value. The idea about the nucleus decaying into three large fragments, of approximately equal masses, arose soon after 1938. At that time, O. Hahn and F. Strassmann discovered the phenomenon of nuclear fission into two fragments [1]. Up to now this phenomenon has not been investigated fully and it deserves further study. In 1941 based on the liquid drop model, R. D. Present in Ref. [2] showed, that increasingly heavier nuclei, with $Z^2/A > 30$, multi-fragment fission (three or more fragments) were more frequent than binary fission. Moreover, heavy nuclei breaking into three or more fragments would release more energy. This phenomena deserves more attention, and experimental information on the fission mechanism would provide unique data to investigate and test laws of formation of nuclei, in particular, to test it near the gap and furthermore the properties of nuclear matter under extreme conditions. Various aspects related to the binary fission are already well understood for different nuclei [3]. Notwithstanding, the mass distribution of

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the ternary fission has not been studied in details heretofore. From the theoretical point of view, different model representations are involved in the interpretation of experimental data on ternary fission [4, 5]. At excitation energies $E^* \sim 3$ MeV/nucleon, ternary decays of the excited nucleus have been accounted for the result of two successive independent acts of binary decay, called cascade fission [6-10]. In such process, light and very heavy fragments can be formed, where the heavy ones can split into another heavy and medium mass fragments still. Likewise, at the same excitation energies, there is an indication [11, 12] of the existence of three-body correlations among the fragments, evincing short time intervals between the decays. Frequently, under a ternary fission, the emission of the light charged particles accompanies fission [9, 10, 13]. Ternary fission can be considered a form of nuclear disintegration into three fragments of a particular state, in which the nucleus is located near the scission point [14]. Alternatively, in the literature it is called cluster decay [15-17]. In the process of nucleus deformation at certain critical neck radius ($\sim 2\text{-}2.5$ fm), the system becomes unstable, where part of the neck is a light particle accompanying fission [18]. After the rupture, a triple nucleus system consisting of fragments and light particle(s) is formed. However, the description of such mechanism still lacks, so a more complete and global theoretical interpretation is needed. Numerous experimental studies have confirmed the existence of ternary fragment in the spontaneous [19-23] and low energy induced fission [24-26], as well as in the fission of actinide and preactinide nuclei induced by incidence of nucleons [27-29], deuterons [30], and heavy ions [7, 9, 12, 28, 31-33], at intermediate energies. Ternary fission is a rare process and the cross-sections for emission of nuclei heavier than α -particles increases when heavier targets are considered. Ternary fission processes can account for several percent of the total fission cross-sections.

The investigation about fission followed by emission of charged particles or light fragments, as well as the measurement and analysis of their mass distribution, energy, and angular distributions, provides additional opportunities to understand the fission mechanism. Furthermore, it brings prominent information about cross-sections and spectra of light nuclei production during ternary fission. The importance and relevance such results are undeniable. They can be used to test and develop nuclear reactions models, as well as in the development of ion sources for nuclear beams. In addition, the new values obtained for the cross-sections can be included in database for reactions induced by nucleons and ions at intermediate energies. It should be remarked that the development of nuclear technology in this field has also stimulated the investigation on some important practical questions. For instance, the investigation of fission induced by particles of different types has stimulated the study of radiation resistance of materials and some other valuable phenomena.

2 Experimental Procedure

A pure natural uranium target of 0.164 g and 0.0487 mm thick was exposed to an accelerated proton beam of 660 MeV in energy from the LNR Phasotron, Joint Institute for Nuclear Research (JINR), Dubna, Russia [34]. The proton flux was determined by the use of an Al monitor [35]. The monitor was irradiated with uranium target and had the same size as the target. The cross-section for the $^{27}\text{Al}(\text{p},3\text{pn})^{24}\text{Na}$ reaction equals 10.8 ± 0.7 mb at 600 MeV of proton energy was used. The irradiation time was 27 min at an proton beam intensity of about 3×10^{14} protons per min.

The cross-sections of radioactive fission fragment production were measured in off-line mode, using a HpGe semiconductor detector. The energy resolution, obtained for the ^{60}Co 1332 keV γ -transition, was 0.13%. The HpGe detector consisted of a $58 \times 40 \times 29$ cm 3 crystal immerse in a cryostat, which was placed in a cubic lead shield. The 7.5 cm thick lead shield wall was covered by a 1 mm cadmium-copper alloy. The energy-dependent detection efficiency of the HpGe detector was up to 10%, and was determined with standard calibration sources of ^{22}Na , ^{54}Mn , $^{57,60}\text{Co}$, and ^{137}Cs . The identification of the reaction products, and the determination of their cross-section production, were performed using data from [36] by means of half-lives, energies and intensities of γ -transition of radioactive fragments.

In the absence of a parent isotope, the cross-section of fission fragment production for each fragment is determined using the following equation:

$$\sigma = \frac{\Delta N \lambda}{N_p N_n k \epsilon \eta (1 - \exp(-\lambda t_1)) \exp(-\lambda t_2)(1 - \exp(-\lambda t_3))}, \quad (1)$$

where σ denotes the cross-section of the reaction fragment production (mb); ΔN is the area under the photopeak; N_p is the proton beam intensity (min $^{-1}$); N_n is the number of target nuclei (in 1/cm 2 units); t_1 is the irradiation time; t_2 is the time of exposure between the end of the irradiation and the beginning of the measurement; t_3 is the time measurement; λ is the decay constant (min $^{-1}$); η is the intensity of γ -transitions; k is the total coefficient of γ -ray absorption in target and detector materials, and ϵ is the γ -ray-detection efficiency.

Usually, the cross-section of an isotope production in the reaction under investigation is direct and independent (I) of the parent nuclei decay, and the cross-section is determined by the eq. (1). If the yield of a given isotope

receives a contribution from the β^\pm -decay of neighboring unstable isobars, the cross-section calculation becomes more complicated [37].

If the formation probability for the parent isotope is known from experimental data or if it can be estimated on the basis of other sources, then the independent yields of daughter nuclei can be calculated by the relation:

$$\sigma_B = \frac{\lambda_B}{(1 - \exp(-\lambda_B t_1)) \exp(-\lambda_B t_2)(1 - \exp(-\lambda_B t_3))} \times \\ \left[\frac{\Delta N}{N_\gamma N_n k \epsilon \eta} - \sigma_A f_{AB} \frac{\lambda_A \lambda_B}{\lambda_B - \lambda_A} \left(\frac{(1 - \exp(-\lambda_A t_1)) \exp(-\lambda_A t_2)(1 - \exp(-\lambda_A t_3))}{\lambda_A^2} \right. \right. \\ \left. \left. - \frac{(1 - \exp(-\lambda_B t_1)) \exp(-\lambda_B t_2)(1 - \exp(-\lambda_B t_3))}{\lambda_B^2} \right) \right], \quad (2)$$

where the subscripts A and B label variables referring to, respectively, the parent and the daughter nucleus; the coefficient f_{AB} specifies the fraction of A nuclei decaying to a B nucleus (this coefficient provides the idea of how much the β -decay affects our data; and $f_{AB} = 1$, when the contribution from the β -decay corresponds 100%); and ΔN is the total photo peak area associated with the decays of the daughter and parent isotopes. The effect of the forerunner can be negligible in some limit cases – for example, in the case where the half-life of the parent nucleus is very long, or in the case where the fraction of its contribution is very small. In the case when parent and daughter isotopes could not be separated experimentally, the calculated cross-sections are classified as cumulative ones (C).

3 Experimental Results

According to the existing theoretical models of fission, we might consider the fragments of mass numbers 7-70 amu, measured in this work, as a third particle accompanying two other fragments. The ternary fragment mass distribution can evince information about the nucleon composition of the neck. Instead, due to the high energy of the fissioning nucleus, the growth of the cross-sections could lead to a partial overlap between the ternary and binary fission, in the mass region of fission fragments 50-70 amu.

The cross-sections of fragment production in ternary fission of uranium induced by 660 MeV proton beam are presented in Table 1. In total, 51 cross-sections are calculated in the fragment mass region $7 < A < 70$ amu. The quoted errors in determining yields received contributions from those associated with the statistical significance of experimental results ($\leq 2\text{-}3\%$), those in measuring the target thickness ($\leq 3\%$), and those in determining the detector efficiency ($\leq 10\%$).

Table 1. The cross-sections of ternary fission fragment production.

Element	Type	Cross-section, mb
⁷ Be	I	1.59±0.16
²² Na	C	0.02±0.002
²⁴ Na	C	0.002±2.0E-4
²⁸ Mg	C	0.0043±4.3E-4
^{34m} Cl	I	7.7E-4±1.5E-5
³⁸ S	I	0.007±1.4E-4
³⁸ Cl	I	≤0.0014
³⁹ Cl	I	0.053±0.005
⁴¹ Ar	C	0.0037±7.4E-4
⁴² K	C	0.007±7.0E-4
⁴³ K	C	0.023±0.002
⁴³ Sc	C	0.012±0.001
⁴⁴ Ar	I	≤0.031
⁴⁴ K	I	2.5E-4±5.0E-5
^{44g} Sc	I	≤0.0025
^{44m} Sc	I	0.065±0.007
⁴⁵ K	C	≤1.3E-4
⁴⁷ Ca	I	0.024±0.002
⁴⁷ Sc	I	0.17±0.02
⁴⁸ Sc	I	0.044±0.004
⁴⁸ V	I	0.022±0.002
⁴⁸ Cr	I	0.014±0.002
⁴⁹ Cr	C	0.025±0.005
⁵¹ Cr	C	0.41±0.04
^{52g} Mn	C	0.0015±1.5E-4
^{52m} Mn	I	0.0085±8.5E-4
⁵² Fe	I	6.5E-4±5.5E-5
⁵⁴ Mn	I	0.11±0.01
⁵⁵ Co	C	0.02±0.002
⁵⁶ Mn	C	0.0053±5.3E-4
⁵⁶ Co	I	0.0022±2.2E-4
⁵⁶ Ni	I	0.063±0.006
⁵⁷ Co	I	0.059±0.006
⁵⁷ Ni	I	0.0011±1.1E-4
^{58(m+g)} Co	I	0.027±0.003
⁵⁹ Fe	C	0.27±0.03
^{60(m+g)} Co	I	0.33±0.03
⁶⁰ Cu	C	≤3.8E-4
⁶¹ Cu	C	0.04±0.004
⁶² Zn	C	0.014±0.001
⁶⁵ Ni	I	0.0017±1.7E-4
⁶⁵ Zn	I	0.10±0.01
⁶⁵ Ga	C	≤0.02
⁶⁶ Ni	I	0.045±0.005
⁶⁶ Ga	I	0.0051±5.1E-4

Table 1 (continuation). The cross-sections of ternary fission fragment production.

Element	Type	Cross-section, mb
$^{46(m+g)}\text{Sc}$	I	0.036 ± 0.004
^{66}Ge	I	0.084 ± 0.008
^{67}Cu	C	0.55 ± 0.06
^{67}Ga	C	0.06 ± 0.006
^{69m}Zn	I	0.041 ± 0.004
^{69}Ge	C	$0.003 \pm 3.0\text{E-}4$

Table 2. The cross-sections for binary (σ_{Bf}) and ternary fission (σ_{Tf}), as well as the ratio $\sigma_{\text{Tf}}/\sigma_{\text{Bf}}$.

Target	σ_{Bf} , mb	σ_{Tf} , mb	$\sigma_{\text{Tf}}/\sigma_{\text{Bf}}$
^{238}U	1226.5 ± 183.9		
	1110 ± 300 [39]	4.43 ± 0.66	0.0036
	1040 ± 75 [40]	0.6 [27]	0.0005 [27]
	1204 ± 133 [40]		

The total cross-section of binary fission (σ_{Bf}) [38], considering the formation of two fragments in one event of fission, the ternary fission cross-section (σ_{Tf}), and the ratio of $\sigma_{\text{Tf}}/\sigma_{\text{Bf}}$ are shown in Table 2. One can see from Table 2 that the binary fission cross-section (σ_{Bf}) in the present work is in good agreement with data from [39, 40] in the same region of the proton energy (E_p). The data for the ternary fission cross-section (σ_{Tf}) at $E_p = 600$ MeV from [27] is smaller than the value of the present work. It should be mentioned that the data for the ternary fission with proton at 600 MeV in ref. [27] is poorly grounded statistically. Notwithstanding, already at 2 GeV proton energy [27] there is good statistics, and the cross-section for light fission fragments production increases. The value for the binary fission in the present work is in good agreement with those from ref. [27] at 600 MeV within statistical errors.

The obtained cross-sections of fragment production from binary and ternary fission of uranium are presented in Fig. 1. Analysis of the binary fission fragment mass distribution is based on the concept of multi-modal fission, described in details in [41]. According the multi-modal fission approach, the total mass-yield distribution of fissioning fragments can be described by three distinct fission modes for the heavy nuclei: symmetric (Superlong) mode and two asymmetric modes – Standard I and Standard II. Superlong mode fragments are strongly elongated, with masses around $A_f/2$, where A_f is the fissioning nucleus mass. Standard I mode is characterized by the influence of the spherical neutron shell $N_H \sim 82$ and proton shell $Z_H \sim 50$, in the heavy fragments with masses $M_H \sim 132\text{-}134$. Standard II mode is characterized by the influence of the deformed neutron shell closure $N_H \sim 86\text{-}88$ and proton shell $Z_H \sim 52$, in the heavy fragments with masses $M_H \sim 138\text{-}140$. From Fig. 1 one can realize that, although the fission cross-sections of light nuclei production with mass $A > 20$ are small, they are cognizable from the mass distribution of binary fission fragments. The present calculations can be considered in the frame of the hypothesis [42, 43] on the light nuclei as a source of ternary fission. This is indicated in Fig. 1, with an extended region of mass from the binary fragment mass distribution.

Systematic studies on ternary fission have been conducted for thermal neutron-induced reactions on ^{249}Cf and ^{235}U targets [18, 44, 45]. Results from [18, 44] for these two reactions show, that for the ^{235}U target the heaviest ternary particle to be observed at the limit of detection was ^{22}O , and for the ^{249}Cf target the heaviest isotopes were ^{37}Si and ^{37}S . There was an indication of a gap in mass number between the heaviest ternary particle and the lightest binary fission fragments, due to low excitation energy. According to the authors, the yield curves are not leveling off, and heavier ternary particles, with still lower yields, are expected to be produced as well. On the other hand, at low energy fission, the masses of ternary particles increase as the mass of the fissioning nucleus [18, 44]. Furthermore, the detailed study regarding the ternary fission in [45] shows an existence of two distinct peaks in mass-yield distribution, corresponding to the light fragments mass 30 and 50-60 amu.

The excitation energy is well known to play an important role on fission dynamics. In low energy fission of actinides, asymmetric fission is controlled by shell effects associated to the fissioning nucleus and fragments [46]. Within the cascade-evaporation model, the projectile energy increasing leads to a higher excitation energy [47], and accordingly, the number of emitted nucleons prior to scission is increased. For heavy target nuclei, prefission neutrons constitute the bulk of emitted nucleons (the emission of protons is strongly suppressed). As the excitation energy of the fissioning nucleus is raised, the gap is rapidly filled, and the ternary and binary yield distributions are smoothly joined. The mean mass of the fissioning nucleus at given proton energy was obtained in our measurements [38] $\bar{A} \sim 227$ amu

and charge $\bar{Z} \sim 92$, corresponding to neutron-deficit uranium isotopes. According to [48], the probability of fission at intermediate energies has a broad distribution versus the number of emitted neutrons. In these cases new fission channels are opened, and hence there is a set of various fissioning nuclides with broad distribution of the excitation energy. This can explain the strong rise of the light fragment formation in the mass range 40-70 amu, at intermediate proton energy.

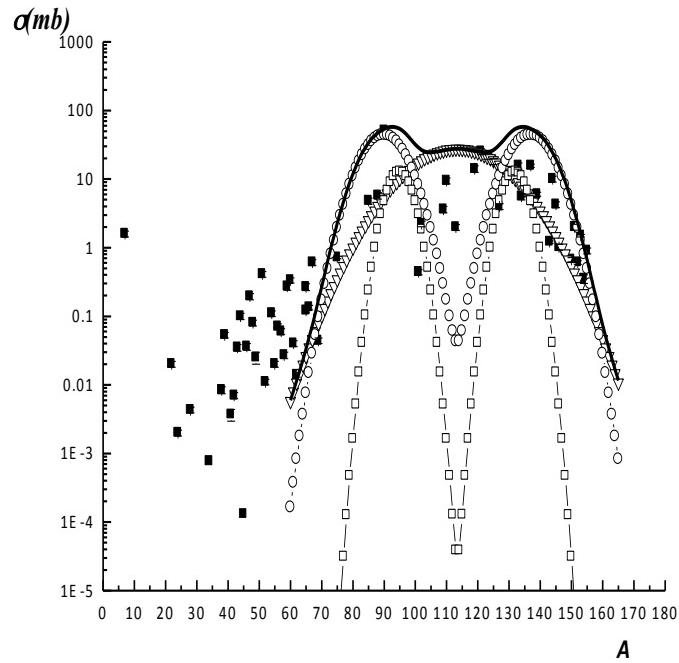


Fig. 1. The mass-yield components of fission fragments for ^{238}U binary fission [38]: Superlong (∇), Standard I (\square), Standard II (\circ), the total fission cross-section (thick black continuous curve). The experimental data (\blacksquare) for binary fission is plotted, concerning the data in [34]. Experimental data of the present work, explicitly evinced in the Table 1 above, are plotted in the interval $A < 70$ amu.

4 Conclusion

The absolute values of 51 cross-sections of fragment production in proton-induced fission of ^{238}U at 660 MeV, which cover the fragment mass region $7 < A < 70$ amu, are determined. We can consider such fragments as a result of nuclear disintegration into three fragments of a particular state, in which fissioning nucleus is located near the scission point. Our experimental data in the present work, concerning ternary fission, complete the ones obtained for binary fission in [34], as illustrated in Fig. 1, as well as the decomposition of the mass-yield distribution from [38]. The cross-section for ternary fission ($\sigma_{\text{Tf}} = 4.43 \pm 0.66$ mb), and the ratio ($\sigma_{\text{Tf}}/\sigma_{\text{Bf}} = 0.0036$) of binary/ternary fission are calculated. Our results are in full compliance with data from [39, 40] in the same region of the proton energy for the binary fission. There is a discrepancy for ternary fission cross-sections in this work and the data from [27] at the same projectile energy, due to the poor statistics used in [27]. A sharp growth of the fragment formation in the mass range 40-70 amu has been found. We can conclude that the probability of finding three fragments is rare, and ternary fission with a large dispersion of masses (two heavy fragments and one lighter) is important and characteristic of intermediate-energy fission. The mass determination of such ternary events is in progress and will contribute to the understanding of fission mechanism.

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